

Europäisches Patentamt  
European Patent Office  
Office européen des brevets



(11) **EP 0 376 052 B1**

(12)

## EUROPEAN PATENT SPECIFICATION

(45) Date of publication and mention  
of the grant of the patent:  
13.03.1996 Bulletin 1996/11

(51) Int Cl.<sup>6</sup>: **C08L 83/10**, **C08L 69/00**,  
**C08K 7/14**  
// (**C08L83/10**, **69:00**),  
(**C08L69/00**, **83:10**)

(21) Application number: **89122982.5**

(22) Date of filing: **13.12.1989**

### (54) Polycarbonate-based resin composition

Harzzusammensetzung auf Basis von Polycarbonat

Composition résineuse à base de polycarbonate

(84) Designated Contracting States:  
**BE CH DE FR GB IT LI NL SE**

(30) Priority: **26.12.1988 JP 326349/88**

(43) Date of publication of application:  
**04.07.1990 Bulletin 1990/27**

(73) Proprietor: **IDEMITSU PETROCHEMICAL CO.**  
**LTD.**  
**Tokyo 100 (JP)**

(72) Inventor: **Okamoto, Masaya**  
**IDEMITSU PETROCHEMICAL CO., LTD.**  
**Ichihara-shi Chiba-ken (JP)**

(74) Representative: **Türk, Gille, Hrabal, Lelfert**  
**Brucknerstrasse 20**  
**D-40593 Düsseldorf (DE)**

(56) References cited:  
**EP-A- 0 193 757** **EP-A- 0 278 498**  
**WO-A-80/00084** **WO-A-91/00885**  
**US-A- 3 640 943** **US-A- 4 224 215**  
**US-A- 4 616 042**

Note: Within nine months from the publication of the mention of the grant of the European patent, any person may give notice to the European Patent Office of opposition to the European patent granted. Notice of opposition shall be filed in a written reasoned statement. It shall not be deemed to have been filed until the opposition fee has been paid. (Art. 99(1) European Patent Convention).

**EP 0 376 052 B1**

## Description

## BACKGROUND OF THE INVENTION

5 Field of the Invention

The present invention relates to a polycarbonate-based resin composition and, more particularly, it relates to a glass fiber-reinforced polycarbonate-based resin composition excelling especially in impact resistance and so suitable for use in various industrial materials.

10

Prior Art

Polycarbonate resins excel in mechanical strength, electrical characteristics, transparency, etc., and find various applications in the form of engineering plastics.

15

Additionally, there is known a glass fiber-reinforced polycarbonate resin in which glass fibers are incorporated into a polycarbonate resin to improve its rigidity and dimensional stability. However, the addition of glass fibers tends to give rise to a drop of Izod impact strength due to brittle fracture.

For that reason, compositions comprising such glass fiber-reinforced polycarbonate resins and further including an organopolysiloxane have been proposed (see JP-B-35929/1984 and JP-A-501860/1982).

20

A problem with molded or otherwise shaped articles obtained from such compositions is, however, that they are poor in insulating properties.

With that problem in mind, compositions of glass fiber-reinforced polycarbonate resins containing a small amount of organopolysiloxane/polycarbonate copolymers have been proposed (see JP-A-160052/1980 and US-A-3 640 943).

25

While such compositions are improved in terms of impact resistance to some extent, their impact resistance is still insufficient to find use in fields for which especially high impact resistance is needed, for instance, chassis or electrically powered tool fields.

Therefore, the object of the present invention is to provide a polycarbonate-based resin composition having a further improved impact resistance.

30

## SUMMARY OF THE INVENTION

As a result of extensive and intensive studies made to provide a solution to the above problems, it has been found that a polycarbonate-based resin composition excelling in not only impact resistance but also rigidity, dimensional stability, etc. can be obtained by using a polycarbonate/polyorganosiloxane copolymer, glass fibers and a polycarbonate resin at a specific proportion. Such findings underlie the present invention.

35

More specifically, the present invention provides a polycarbonate-based resin composition, comprising (each based on the composition)

40

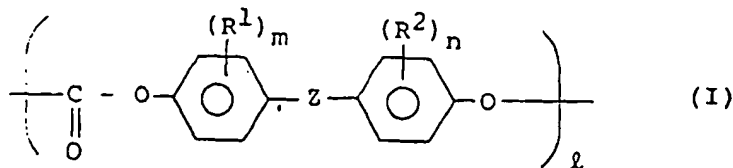
10 to 80 % by weight of a polycarbonate/polyorganosiloxane Copolymer having a viscosity average molecular weight (Mv) of from 10 000 to 40 000,

20 to 50 % by weight of glass fibers, and

45

0 to 70 % by weight of a polycarbonate resin, which is a polycarbonate homopolymer or copolymer obtained by the reaction of at least one kind of divalent phenols and phosgene and consisting of repeating units expressed by the following formula (I):

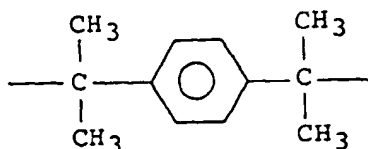
50



55

wherein:

Z is a single bond, an ether bond, an alkylene group having 1 to 8 carbon atoms, an alkylidene group having 2 to 8 carbon atoms, a cycloalkylene group having 5 to 15 carbon atoms, a cycloalkylidene group having 5 to 15 carbon atoms, a sulfonyl group, a sulfoxide group, a carbonyl group, a sulfide group or a group:



$R^1$  and  $R^2$ , which may be identical with or different from each other, each stand for a hydrogen atom, a halogen atom or an alkyl group having 1 to 8 carbon atoms,  
 $m$  and  $n$  each stand for an integer of 1 to 4, provided that when  $m$  is 2 or more,  $R^1$  may be identical or different and when  $n$  is 2 or more,  $R^2$  may be identical or different, and  
 $l$  is between 3 and 50,

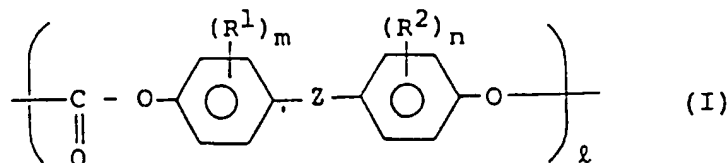
the amount of said polyorganosiloxane accounting for 3.5 to 29 % by weight of said resin components.

The claimed resin composition defined above is characterized by having not only a further improved impact resistance but also a high elastic modulus and bending strength.

#### DETAILED DESCRIPTION OF THE INVENTION

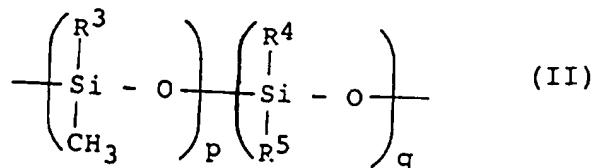
According to a preferred embodiment of the present invention the polycarbonate/polyorganosiloxane copolymer used in the present invention is comprised of

a polycarbonate segment having repeating units expressed by the following formula (I):



wherein Z,  $R^1$ ,  $R^2$ ,  $m$ ,  $n$  and  $l$  having the meanings as defined above, and

a polyorganosiloxane segment having repeating units expressed by the following formula (II):



wherein:

$R^3$ ,  $R^4$  and  $R^5$ , which may be identical with or different from one another, each stand for a hydrogen atom, an alkyl group having 1 to 6 carbon atoms or a phenyl group, and  
 $p$  and  $q$  each are an integer of 1 or more.

Usually, the polyorganosiloxane segment has a degree of polymerization of 5 to 300.  
 According to further preferred embodiments of the present invention

the amount of the polycarbonate/polyorganosiloxane copolymer is 15 to 80 % by weight of the composition, and

the glass fibers used according to the present invention preferably are surface-treated.

5 The above polycarbonate/polyorganosiloxane copolymer is a block copolymer comprising the polycarbonate segment having repeating units expressed by the general formula (I) and the polyorganosiloxane segment having repeating units expressed by the general formula (II), and has a viscosity-average molecular weight of 10,000 to 40,000, preferably 15,000 to 35,000.

10 For instance, such a polycarbonate/polyorganosiloxane copolymer may be prepared by dissolving a pre-prepared polycarbonate oligomer forming the polycarbonate segment and a polyorganosiloxane having a terminal reactive group and forming the polyorganosiloxane segment in a solvent such as methylene chloride, chlorobenzene or pyridine, adding an aqueous sodium hydroxide solution of bisphenol to the resulting solution and subjecting that solution to an interface reaction with a catalyst such as triethylamine or trimethylbenzyl-ammonium chloride. Use may also be made of polycarbonate/polyorganosiloxane copolymers prepared by such methods as set forth in JP-B-30108/1969 and 20510/1970.

15 The polycarbonate having repeating units expressed by the general formula (I) may be prepared by a solvent method in which a divalent phenol is allowed to react with a carbonate precursor such as phosgene or subjected to an ester exchange reaction with a carbonate precursor such as diphenyl carbonate in a solvent such as methylene chloride in the presence of an acid acceptor and a molecular-weight regulator, both known in the art.

20 The divalent phenols preferably used in the present invention include bisphenols. Particular preference is given to 2,2-bis(4-hydroxyphenyl) propane (bisphenol A). The bisphenol A may be partly or wholly substituted by other divalent phenols. As the divalent phenols other than the bisphenol A, reference may be made to compounds such as, for instance, (4-hydroxyphenyl) alkanes, hydroquinone, 4,4'-dihydroxy-diphenyl, bis(4-hydroxyphenyl) cycloalkanes, bis(4-hydroxyphenyl) sulfide, bis(4-hydroxyphenyl) sulfone, bis(4-hydroxyphenyl) sulfoxide, bis(4-hydroxyphenyl) ether and bis(4-hydroxyphenyl) ketone or halogenated bisphenols such as bis(3,5-dibromo-4-hydroxyphenyl) propane and bis(3,5-dichloro-4-hydroxyphenyl) propane.

25 The polycarbonate may be a homopolymer (oligomer) with one of such divalent phenols or a copolymer with two or more thereof. Alternatively, it may be a thermoplastic branched polycarbonate obtained by using the above divalent phenols in combination with a polyfunctional aromatic compound.

30 For instance, the polyorganosiloxane expressed by the general formula II may be obtained by the reaction of a dialkyldichlorosilane and/or diaryldichlorosilane with water. The monomer of such a polyorganosiloxane, may include dimethylsiloxane or methylphenylsiloxane.

35 Such a polycarbonate/polyorganosiloxane copolymer as mentioned above is incorporated into the composition at a proportion of 10 to 80% by weight, preferably 15 to 80% by weight. If the quantity of said copolymer is less than 10% by weight, there is then no improvement in impact resistance. In a quantity higher than 80% by weight, on the other hand, there is a drop of dimensional stability.

The quantity of the polyorganosiloxane contained in the resin except the glassy component (a total quantity of the polycarbonate/polyorganosiloxane copolymer + the polycarbonate resin) is 3.5 to 29% by weight. If the quantity of the polyorganosiloxane is below 3.5% by weight, there is then no improvement in impact resistance. In a quantity exceeding 29% by weight, on the other hand, any copolymer having sufficient molecular weight cannot be obtained.

40 Referring then to the glass fibers used in the present invention, any one of alkali-containing glass, low-alkali glass and non-alkali glass may be used to this end. Preferably, the glass fibers used are 1 to 8 mm, particularly 3 to 6 mm in length and 3 to 20  $\mu$ m, particularly 5 to 15  $\mu$ m in diameter. The glass fibers may be used in any nonrestrictive forms such as rovings, milled fibers and chopped strands, and may be used alone or in combination of two or more.

45 As such glass fibers, use may also be made of those surface-treated with silane coupling agents such as aminosilanes, epoxysilanes, vinylsilanes or methacrylsilanes, or chromium complex compounds, boron compounds or like other compounds to improve their affinity with respect to the polycarbonate/polyorganosiloxane copolymers and polycarbonate resins.

Such glass fibers as mentioned above are incorporated into the composition at a proportion of 20 to 50% by weight. A proportion of the glass fibers less than 20% by weight is unpreferred, since there is then a drop of dimensional stability. A proportion of the glass fibers exceeding 50% by weight is again unpreferred, since kneading is unfeasible.

According to the present invention, a polycarbonate resin may be used, if required.

The polycarbonate resin has repeating units expressed by the general formula (I) and, as mentioned above, may be easily obtained by the reaction of a divalent phenol with phosgene by way of example. The divalent phenols used may include such phenols as already mentioned.

55 The polycarbonate resin used in the present invention has a viscosity-average molecular weight of from 10,000 to 40,000, preferably 20,000 to 40,000.

The polycarbonate resin is incorporated into the composition at a proportion of 10 to 80% by weight. If the proportion of the polycarbonate resin exceeds 80% by weight, there is then no improvement in impact resistance.

The polycarbonate-based resin composition according to the present invention is essentially comprised of the above polycarbonate/polyorganosiloxane copolymer, glass fibers and polycarbonate resin, and may additionally contain any various additives, if required, provided that the object of the present invention is achievable. For instance, carbon fibers, metal fibers, inorganic fillers, metal powders, UV absorbers, flame retardants, and colorants may be added.

The polycarbonate-based resin composition according to the present invention may be obtained by blending and kneading the above components together. To this end, blending and kneading may be carried out in conventional manners with, for instance, ribbon blenders, Henschel mixers, Banbury mixers, drum tumblers, single-, twin- or multi-screw extruders, Ko-kneaders or like other equipment. For kneading, a heating temperature of 250 to 300°C is usually applied.

The polycarbonate-based resin composition according to the present invention excels in not only impact resistance but also rigidity expressed in terms of bending strength and tensile modulus of elasticity or dimensional stability.

Thus, the present compositions are effectively used in various industrial fields inclusive of electric/electronic fields, in particular fields for which high impact resistance is needed, e.g., chassis or like other fields.

## EXAMPLES

The present invention will now be explained in more detail with reference to the following examples.

### Preparation Example 1-1 (Preparation of Reactive Polydimethylsiloxane)

Over two hours, a mixture of 100 g of water with 206 g of dioxane was added to 800 g of dimethyldichlorosilane. Under mild reflux, the resulting mixture was heated, while stirred, into a homogeneous state. The mixture was stripped in vacuo and brought up to 202°C at a pressure of 1.6 kPa (12 mmHg). Then, the stripped product was filtrated to obtain a transparent oily product. Dissolved in 130 g of dry dichloromethane were 225 g of such an oily product, and the solution was added under intensive agitation to a mixture of 114 g of bisphenol A, 130 g of dry pyridine and 1300 g of dichloromethane over 65 minutes. Afterwards, the product was alkali-washed with 1000 g of an aqueous solution of sodium hydroxide (0.01N), then acid-washed with 1000 g of hydrochloric acid (0.1N) and finally washed with 1000 g of water. Removal of dichloromethane by evaporation gave the end reactive polydimethylsiloxane (reactive PDMS for short) having a terminal phenolic hydroxyl group.

### Preparation Example 1-2 (Preparation of Reactive PDMS)

In Preparation Example 1-1, the amount of the first-mentioned water was changed from 100 g to 140 g. Under otherwise similar conditions, the reactive PDMS was obtained.

### Preparation Example 1-3 (Preparation of Reactive PDMS)

As the reactive PDMS, use was made of silicone oil reactive at both terminals (KF 6002®, Shinetsu Silicone Co., Ltd.).

### Preparation Example 1-4 (Preparation of Reactive PDMS)

Mixed together were 1483 g of octamethylcyclotetrasiloxane, 137 g of 1,1,3,3-tetramethyldisiloxane and 35 g of 86% sulfuric acid, and the mixture was stirred at room temperature for 17 hours. Afterwards, an oily phase was separated, and 25 g of sodium hydrogencarbonate were added thereto, followed by one-hour stirring. After filtration, the product was distilled in vacuo at 150°C and 400 Pa (3 mmHg) to remove low-boiling matters.

Added to a mixture of 60 g of 2-allylphenol with 0.0014 g of platinum in the form of a platinum/alcolate complex were 294 g of the above-obtained oil at a temperature of 90°C. The mixture was stirred for 3 hours, while maintained at a temperature of 90°C to 115°C. The product was extracted with methylene chloride and washed with three portions of 80% aqueous methanol to remove excessive 2-allylphenol. The product was dried over anhydrous sodium sulfate and rid of the solvents in vacuo at a temperature of up to 115°C to obtain the reactive PDMS.

### Preparation Example 1-5 (Preparation of Reactive PDMS)

As the reactive PDMS, use was made of silicone oil reactive at both terminals (X-22-165B®, Shinetsu Silicone Co., Ltd.).

Preparation Example 1-6 (Preparation of Reactive PDMS)

As the reactive PDMS, use was made of silicone oil reactive at both terminals (X-22-165®, Shientsu Silicone Co., Ltd.).

Preparation Example 2 (Preparation of Polycarbonate Oligomer)

Sixty (60) kg of bisphenol A were dissolved in 400 liters of a 5% aqueous solution of sodium hydroxide to prepare an aqueous solution of bisphenol A in sodium hydroxide. Then, the solution was maintained at room temperature. The aqueous sodium hydroxide solution of bisphenol A and methylene chloride were introduced at the respective flow rates of 138 liters per hour and 69 liters per hour into a tubular reactor of 10 mm in inner diameter and 10 m in length through an orifice plate, into which 10.7 kg/hour of phosgene were blown in cocurrent relation thereto, for three-hour continuous reactions. The tubular reactor used was of a double structure designed to pass cooling water through an outer jacket to keep the discharge temperature of the reaction solution at 25°C. The discharge solution was also regulated to pH 10-11. The thus obtained reaction solution was allowed to stand to separate and remove an aqueous phase, thereby obtaining a methylene chloride phase (220 liters), to which 170 liters of methylene chloride were added under sufficient agitation to obtain a polycarbonate oligomer (with a concentration of 317 g/liters). This polycarbonate oligomer (PC oligomer for short) was found to have a degree of polymerization of 3-4.

Preparation Example 3-1 (Preparation of PC-PDMS Copolymer A)

Dissolved in 2 liters of methylene chloride were 160 g of the reactive PDMS obtained in Preparation Example 1-1, and the solution was mixed with 10 liters of the PC oligomer obtained in Preparation Example 2. Added to the mixture were 26 g of sodium hydroxide dissolved in 1 liter of water and 5.7 cc of triethylamine, followed by one-hour stirring at 500 rpm and room temperature. Afterwards, 600 g of bisphenol A dissolved in 5 liters of an aqueous solution of 5.2% by weight sodium hydroxide, 8 liters of methylene chloride and 81 g of p-tert.-butylphenol were added to the solution, followed by two-hour stirring at 500 rpm and room temperature. Thereafter, an additional 5 liters of methylene chloride were added to the solution, and the resulting product was washed with 5 liters of water, then alkali-washed with 5 liters of a 0.01 N aqueous solution of sodium hydroxide, then acid-washed with 5 liters of 0.1 N hydrochloric acid and finally washed with 5 liters of water. Subsequent removal of methylene chloride gave a PC-PDMS copolymer A in the form of chips, which found to have a PDMS content of 3.5% by weight.

Preparation Example 3-2 (Preparation of PC-PDMS Copolymer B)

In Preparation Example 3-1, 500 g of the reactive PDMS obtained in Preparation Example 1-2 were used in place of 160 g of the reactive PDMS obtained in Preparation Example 1-1. Under otherwise similar conditions as in Preparation Example 3-1, a PC-PDMS copolymer B was prepared, which was found to have a PDMS content of 10% by weight.

Preparation Example 3-3 (Preparation of PC-PDMS Copolymer C)

In Preparation Example 3-1, 2.6 kg of the reactive PDMS obtained in Preparation Example 1-2 were used in place of 160 g of the reactive PDMS obtained in Preparation Example 1-1 and the amount of sodium hydroxide used was changed from 26 g to 50 g. Under otherwise similar conditions as in Preparation Example 3-1, a PC-PDMS copolymer C was prepared, which was found to have a PDMS content of 29% by weight.

Preparation Example 3-4 (Preparation of PC-PDMS Copolymer D)

Dissolved in 9.5 liters of the PC oligomer obtained in Preparation Example 2 were 480 g of the reactive PDMS obtained in Preparation Example 1-3 (silicone oil reactive at both terminals), and 101 g of triethylamine were slowly added dropwise to the solution under agitation. After the dropwise addition, the solution was stirred for 1 hour and then acid-washed with 5 liters of 0.1 N hydrochloric acid to separate an organic phase. Thereafter, added to the solution were 600 g of bisphenol A dissolved in 5 liters of an aqueous solution of 5.2% by weight sodium hydroxide, 8 liters of methylene chloride and 25 g of p-tert.-butylphenol, followed by two-hour stirring at 500 rpm and room temperature. After that, additional 5 liters of methylene chloride were added to the solution, which was in turn washed with 5 liters of water, then alkali-washed with 5 liters of a 0.01 N aqueous solution of sodium hydroxide, then acid-washed with 5 liters of 0.1 N hydrochloric acid and finally washed with 5 liters of water. Subsequent removal of methylene chloride gave a PC-PDMS copolymer D in the form of chips, which was found to have a PDMS content of 3.9% by weight.

Preparation Example 3-5 (Preparation of PC-PDMS Copolymer E)

As the PC-PDMS copolymer, use was made of Macrolon Type 1207® produced by Bayer Corp., which had a PDMS content of 4.8% by weight.

5 It is understood that the PDMS contents of the above PC-PDMS copolymers were all determined by H NMR.

Preparation Example 3-6 (Preparation of PC-PDMS Copolymer F)

10 In Preparation Example 3-1, the reactive PDMS obtained in Preparation Example 1-4 was used in place of the reactive PDMS obtained in Preparation Example 1-1. Under otherwise similar conditions as in Preparation Example 3-1, a PC-PDMS copolymer F was obtained, which was found to have a PDMS content of 3.5% by weight.

Preparation Example 3-7 (Preparation of PC-PDMS Copolymer G)

15 In Preparation Example 3-1, the reactive PDMS obtained in Preparation Example 1-5 was used in place of the reactive PDMS obtained in Preparation Example 1-1. Under otherwise similar conditions as in Preparation Example 3-1, a PC-PDMS copolymer G was obtained, which was found to have a PDMS content of 3.5% by weight.

Preparation Example 3-8 (Preparation of PC-PDMS Copolymer H)

20 In Preparation Example 3-1, the reactive PDMS obtained in Preparation Example 1-6 was used in place of the reactive PDMS obtained in Preparation Example 1-1. Under otherwise similar conditions as in Preparation Example 3-1, a PC-PDMS copolymer H was obtained, which was found to have a PDMS content of 3.5% by weight.

25 Examples 1-18 and Comparative Examples 1-5

The PC-PDMS copolymers A-H obtained in Preparation Examples 3-1 to 3-8, polycarbonate (having an average molecular weight of 25,000; and available under the trade name of Toughron A-2500, produced by Idemitsu Petrochemical Co., Ltd.) and glass fibers (non-alkali glass surface-treated with aminosilane; and 6 mm in length and 13 µm in diameter) were blended together at the proportions specified in Table 1, and the blends were formed through a 30-mm vented extruder into pellets, which were in turn injection-molded at a temperature of 300°C to obtain molded samples for the determination of their physical properties. The results are set forth in Table 1. It is understood that the glass fibers were supplied downstream of the hopper of the extruder through which the resin stock was fed in.

Table 1

	PC-PDMS Copolymer		Polycarbonate (weight %)	PDMS Content of Resin Components (weight %)	Glass fibers (weight %)	Izod Impact Strength *1 (kg·cm/cm)	Tensile Modulus *2 (kg/cm <sup>2</sup> )	Bending Strength *3 (kg/cm <sup>2</sup> )
	Types	Amount (weight %)						
Example 1	A	50	0	3.5	50	31	84800	2400
" 2	A	70	0	3.5	30	27	58900	1800
Comp.Expl.1	A	90	0	3.5	10	20	33500	1200
" 3	B	70	0	10	30	28	59000	1800
" 4	C	50	0	29	50	30	85200	2300
" 5	C	70	0	29	30	30	59000	1700
Comp.Expl.2	C	90	0	29	10	21	33600	1100
" 6	D	70	0	3.9	30	29	59100	1800
Comp.Expl.3	A	30	60	1.2	10	20	33700	1200
Comparative	-	0	90	0	10	11	33700	1200
Example 4	-	10	70	3.6	20	23	46300	1600
Example 7	C	10	80	0	20	15	46500	1600
Comparative	-	0	50	2.9	30	28	59000	1800
Example 5	B	20	30	5.7	30	27	59200	1800
Example 8	B	40	10	8.6	30	27	58900	1700
" 9	B	60	60	4.1	30	28	59000	1800
" 10	C	10	65	2.1	30	18	58900	1700
" 11	C	5	70	0	30	18	59000	1800
Comparative	C	0	50	0	50	19	85000	2400
Example 6	-	0	70	4.8	30	27	58900	1700
" 7	-	0	0	3.5	30	21	58800	1700
" 8	-	0	0	3.5	30	22	58900	1800
Example 12	E	70	0	3.5	30	23	58800	1700
" 13	F	70	0	3.5	30	22	58900	1800
" 14	G	70	0	3.5	30	23	58800	1700
" 15	H	70	0	3.5	30	23	58800	1700

\*1: Measured according to JIS-K-7110

\*2: Measured according to JIS-K-7113

\*3: Measured according to JIS-K-7203



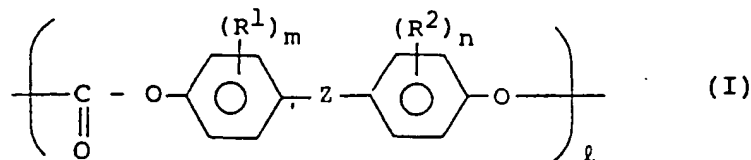
## Claims

## 1. A polycarbonate-based resin composition comprising (each based on the composition)

10 to 80 % by weight of a polycarbonate/polyorganosiloxane copolymer having a viscosity average molecular weight (Mv) of from 10 000 to 40 000,

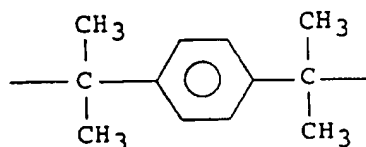
20 to 50 % by weight of glass fibers, and

0 to 70 % by weight of a polycarbonate resin, which is a polycarbonate homopolymer or copolymer obtained by the reaction of at least one kind of divalent phenols and phosgene and consisting of repeating units expressed by the following formula (I):



wherein:

Z is a single bond, an ether bond, an alkylene group having 1 to 8 carbon atoms, an alkylidene group having 2 to 8 carbon atoms, a cycloalkylene group having 5 to 15 carbon atoms, a cycloalkylidene group having 5 to 15 carbon atoms, a sulfonyl group, a sulfoxide group, a carbonyl group, a sulfide group or a group:



R<sup>1</sup> and R<sup>2</sup>, which may be identical with or different from each other, each stand for a hydrogen atom, a halogen atom or an alkyl group having 1 to 8 carbon atoms,

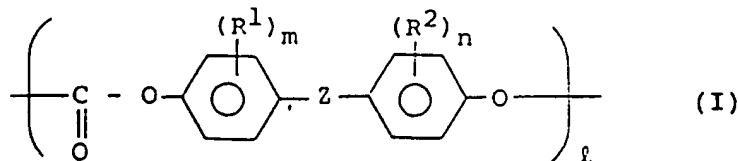
m and n each stand for an integer of 1 to 4, provided that when m is 2 or more, R<sup>1</sup> may be identical or different and when n is 2 or more, R<sup>2</sup> may be identical or different, and

l is between 3 and 50,

the amount of said polyorganosiloxane accounting for 3.5 to 29 % by weight of said resin components.

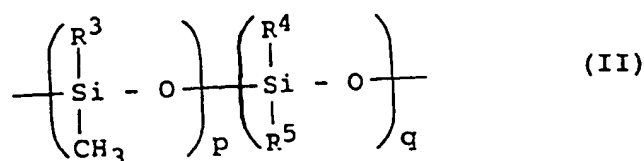
2. The polycarbonate-based resin composition as claimed in claim 1, wherein the polycarbonate/polyorganosiloxane copolymer is comprised of

a polycarbonate segment having repeating units expressed by the following formula (I):



wherein Z, R<sup>1</sup>, R<sup>2</sup>, m, n and l having the meanings as defined in claim 1, and

a polyorganosiloxane segment having repeating units expressed by the following formula (II):



wherein:

$\text{R}^3$ ,  $\text{R}^4$  and  $\text{R}^5$ , which may be identical with or different from one another, each stand for a hydrogen atom, an alkyl group having 1 to 6 carbon atoms or a phenyl group, and  $p$  and  $q$  each are an integer of 1 or more.

3. The polycarbonate-based resin composition as claimed in claim 1 or 2, wherein the amount of the polycarbonate/polyorganosiloxane copolymer is 15 to 80 % by weight of the composition.
4. The polycarbonate-based resin composition as claimed any of claims 1 - 3, wherein the glass fiber is surface-treated.

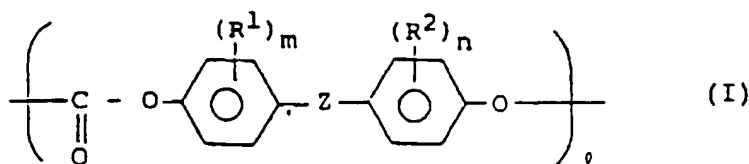
## Patentansprüche

1. Harzzusammensetzung auf Polycarbonatbasis umfassend (jeweils basierend auf der Zusammensetzung)

10 bis 80 Gew.% eines Polycarbonat/Polyorganosiloxancopolymers mit einem durchschnittlichen Viskositätsmolekulargewicht (Mv) von 10.000 bis 40.000,

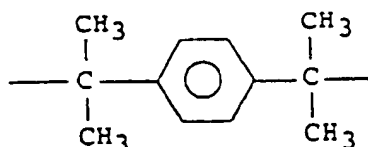
20 bis 50 Gew.% Glasfasern und

0 bis 70 Gew.% eines Polycarbonatharzes, das ein Polycarbonathomopolymer oder -copolymer ist, erhalten durch Reaktion von wenigstens einer Art divalenter Phenole und Phosgene und bestehend aus sich wiederholenden Einheiten, die durch die folgende Formel (I) ausgedrückt werden:



worin:

$\text{Z}$  eine Einfachbindung ist, eine Etherbindung, eine Alkylengruppe mit 1 bis 8 Kohlenstoffatomen, eine Alkylidengruppe mit 2 bis 8 Kohlenstoffatomen, eine Cycloalkylengruppe mit 5 bis 15 Kohlenstoffatomen, eine Cycloalkylidengruppe mit 5 bis 15 Kohlenstoffatomen, eine Sulfonylgruppe, eine Sulfoxidgruppe, eine Carbonylgruppe, eine Sulfidgruppe oder eine Gruppe:



$\text{R}^1$  und  $\text{R}^2$ , die gleich oder verschieden voneinander sein können, können Wasserstoff, Halogen oder Alkyl mit 1 bis 8 Kohlenstoffatomen sein,  $m$  und  $n$  stehen jeweils für eine ganze Zahl von 1 bis 4, vorausgesetzt, daß, wenn  $m$  gleich 2 oder mehr

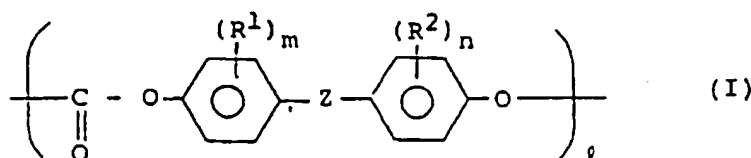
ist, R<sup>1</sup> gleich oder verschieden ist, und wenn n gleich 2 oder mehr ist, R<sup>2</sup> gleich oder verschieden ist, und

I zwischen 3 und 50 ist,

die Menge dieses Polyorganosiloxans ist 3,5 bis 29 Gew.% der besagten Harzkomponente.

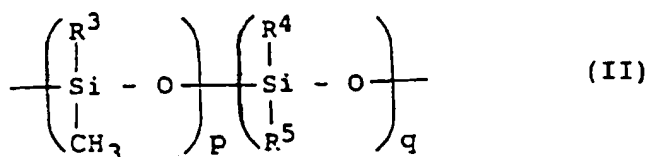
2. Polycarbonatzusammensetzung auf Polycarbonatbasis gemäß Anspruch 1, wobei das Polycarbonat/Polyorganosiloxan-copolymer besteht aus

einem Polycarbonatsegment mit sich wiederholenden Einheiten ausgedrückt durch die nachfolgende Formel (I):



worin Z, R<sup>1</sup>, R<sup>2</sup>, m, n und l dieselbe Bedeutung haben wie vorstehend definiert und

ein Polyorganosiloxansegment mit sich wiederholenden Einheiten ausgedrückt durch die nachstehende Formel (II):



worin:

R<sup>3</sup>, R<sup>4</sup> und R<sup>5</sup>, die identisch oder verschieden voneinander sein können, jeweils für Wasserstoff, eine Alkylgruppe mit 1 bis 6 Kohlenstoffatomen oder eine Phenylgruppe stehen und p und q jeweils eine ganze Zahl von 1 oder mehr sind.

3. Polycarbonatharzzusammensetzung auf Polycarbonatbasis gemäß Anspruch 1 oder 2, wobei die Menge an Polycarbonat/ Polyorganosiloxanocopolymer 15 bis 80 Gew.% der Zusammensetzung ist.
4. Harzzusammensetzung auf Polycarbonatbasis gemäß einem der Ansprüche 1 bis 3, wobei die Glasfaser oberflächenbehandelt ist.

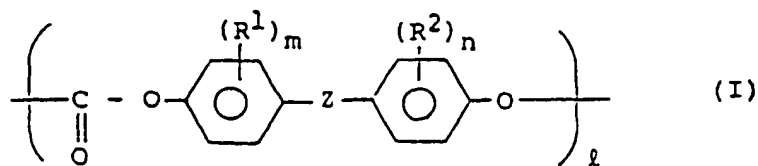
## Revendications

1. Composition de résine à base de polycarbonate comprenant (à chaque fois sur base de la composition)

10 à 80% en poids d'un copolymère de polycarbonate/ polyorganosiloxane possédant un poids moléculaire moyen selon la viscosité (Mv) de 10.000 à 40.000,

20 à 50% en poids de fibres de verre et

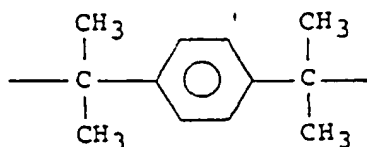
0 à 70% en poids d'une résine de polycarbonate, qui est un copolymère ou un homopolymère de polycarbonate, obtenu par la réaction d'au moins une sorte de phénols divalents et de phosgène et constitué d'unités récurrentes représentées par la formule (I)



dans laquelle

Z

représente une simple liaison, une liaison éther, un radical alkylène comportant de 1 à 8 atomes de carbone, un radical alkylidène comportant de 2 à 8 atomes de carbone, un radical cycloalkylène comportant de 5 à 15 atomes de carbone, un radical cycloalkylidène comportant de 5 à 15 atomes de carbone, un radical sulfonyle, un radical sulfoxyde, un radical carbonyle, un radical sulfure, ou un radical de la formule :



les symboles  $\text{R}^1$  et  $\text{R}^2$ ,

qui peuvent être identiques entre eux ou différer les uns des autres, représentent chacun un atome d'hydrogène, un atome d'halogène, ou un radical alkyle comportant de 1 à 8 atomes de carbone,

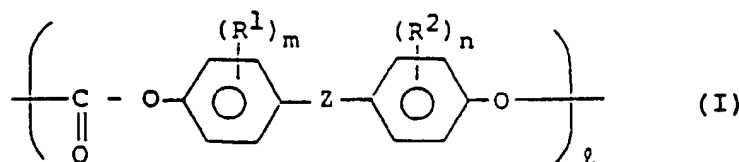
m et n

représentent chacun un nombre entier dont la valeur varie de 1 à 4, avec la condition que lorsque m est égal ou supérieur à 2,  $\text{R}^1$  peut être identique ou différent et que lorsque n est égal ou supérieur à 2,  $\text{R}^2$  peut être identique ou différent et a une valeur qui varie de 3 à 50,

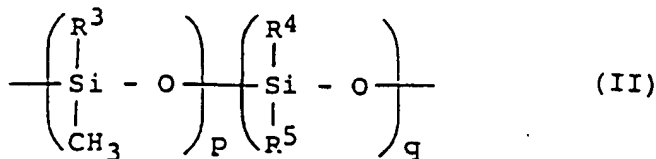
la quantité dudit polyorganosiloxane s'élevant de 3,5 à 29% en poids desdits composants de la résine.

2. Composition de résine à base de polycarbonate suivant la revendication 1, caractérisée en ce que le copolymère de polycarbonate/polyorganosiloxane est constitué de

un segment polycarbonate comportant des unités récurrentes, répondant à la formule (I) qui suit :



dans laquelle Z,  $\text{R}^1$ ,  $\text{R}^2$ , m, n et l possèdent les significations qui leur ont été attribuées dans la revendication 1 et d'un segment de polyorganosiloxane comportant des unités récurrentes répondant à la formule (II) qui suit :



dans laquelle

$R^3$ ,  $R^4$  et  $R^5$  qui peuvent être identiques entre eux ou différer les uns des autres, représentent chacun un atome d'hydrogène, un radical alkyle comportant de 1 à 6 atomes de carbone, ou un radical phényle et p et q représentent chacun un nombre entier égal ou supérieur à 1.

- 5      3. Composition de résine à base de polycarbonate suivant la revendication 1 ou 2, caractérisée en ce que la quantité de copolymère de polycarbonate/polyorganosiloxane varie de 15 à 80% en poids de la composition.
- 10      4. Composition de résine à base de polycarbonate suivant l'une quelconque des revendications 1 à 3, caractérisée en ce que les fibres de verre sont traitées en surface.

15

20

25

30

35

40

45

50

55